2009

Air-Sea Gas Exchange Of Co2 and Dms In the North Atlantic By Eddy Covariance

Scott Miller
State University of New York - Albany

Christa Marandino
University of California - Irvine

Warren J. De Bruyn
Chapman University, debruyn@chapman.edu

Eric S. Saltzman
University of California - Irvine

Recommended Citation

Miller, Scott, et al. "Air-sea gas exchange of CO2 and DMS in the North Atlantic by eddy covariance." *Geophysical Research Letters* 36.15 (2009). doi: 10.1029/2009GL038907

Air-Sea Gas Exchange Of Co2 and Dms In the North Atlantic By Eddy Covariance

Air-sea gas exchange of CO_2 and DMS in the North Atlantic by eddy covariance

Scott Miller,[1] Christa Marandino,[2] Warren de Bruyn,[3] and Eric S. Saltzman[2]

Received 27 April 2009; revised 10 June 2009; accepted 6 July 2009; published 8 August 2009.

[1] We report the first simultaneous eddy covariance flux measurements of CO_2 and dimethylsulfide (DMS) over the open ocean for two North Atlantic cruises. After normalization for Schmidt number, the two gases give essentially identical gas transfer coefficients and wind speed dependences for the wind speed range 2–10 ms^{-1}. The data indicate a linear relationship between the gas transfer coefficient and mean wind speed, with measured gas transfer coefficients slightly above the Wanninkhof (1992) parameterization, particularly at low wind speeds. **Citation:** Miller, S., C. Marandino, W. de Bruyn, and E. S. Saltzman (2009), Air-sea gas exchange of CO_2 and DMS in the North Atlantic by eddy covariance, *Geophys. Res. Lett.*, *36*, L15816, doi:10.1029/2009GL038907.

1. Introduction

[2] Most current parameterizations of air-sea gas exchange utilize a coefficient k (the piston velocity) to relate the gas flux to the air-sea concentration difference. This coefficient incorporates all the physical factors controlling gas transport through both sides of the air-sea interface. For different gases, the effect of molecular diffusivity on k is accounted for by normalizing to a common Schmidt number ($Sc = \upsilon/D$, where υ is the seawater kinematic viscosity and D is the diffusivity of the gas in seawater) via $k\mathrm{Sc}^{1/2} = const$. Tracer release studies [*Ho et al.*, 2006; *Liss and Merlivat*, 1986; *Nightingale et al.*, 2000; *Wanninkhof et al.*, 1985] and wind-wave tank experiments [*Broecker and Siems*, 1984; *Jahne and Haussecker*, 1998] showed that k increases with wind speed, though constraining the functional form of this dependence in the oceans has proven challenging due to the long averaging time of tracer studies, and processes affecting gas exchange that are not correlated to wind speed. Theory, laboratory and modeling studies suggest that gas solubility also affects air-sea gas transfer when whitecaps are present due to bubble-mediated transport [*Woolf*, 1997].

[3] During the past decade, the eddy covariance (EC) technique has been used from ships at sea to measure CO_2 and DMS fluxes and piston velocities on shorter temporal and spatial scales. These data show a wide range of wind speed dependencies for k. For low-moderate wind speed, the Gas Ex 2001 experiment (Gas Ex-2001) [*McGillis et al.*, 2004] showed a very weak relationship for k , while four DMS eddy covariance studies exhibited an approximately linear dependence of k on wind speed [*Marandino et al.*, 2007, 2008a; *Huebert et al.*, 2004; *Blomquist et al.*, 2006]. For higher wind speeds, up to 16 ms^{-1}, the Gas Ex 1998 experiment (Gas Ex-1998) [*McGillis et al.*, 2001] showed a strong (quadratic to cubic) relationship of k versus wind speed. Some of the differences between k during Gas Ex-1998 and Gas Ex-2001 were hypothesized to reflect differences in physical forcing of gas exchange in different environments [*Hare et al.*, 2004], while differences between k_{CO2} and k_{DMS} measured during different cruises have been hypothesized to reflect the different solubilities of these two gases and the importance of bubble-mediated transport [*Blomquist et al.*, 2006].

[4] Here we present EC-based CO_2 and DMS fluxes and piston velocities measured in the North Atlantic during summer 2007. We examine the relationship between the piston velocity and wind speed, and use the gas transfer coefficients of CO_2 and DMS measured on the same cruise to provide insight into the sensitivity of gas exchange to gas solubility during low to moderate wind conditions.

2. Methods

[5] The measurements were made from the research vessel R/V Knorr during cruises in summer 2007 from Bridgetown, Barbados to Reykjavik, Iceland (Knorr-07a, 29 May-9 Jun), and from Reykjavik to Woods Hole, MA (Knorr-07b, 17–26 Jul). Gas transfer coefficients were determined according to $F = k(C_w - C_a/\alpha)$, where F is the turbulent air-sea species flux, C_w and C_a are the bulk seawater and air concentrations, and α is the dimensionless solubility. CO_2 and DMS fluxes were measured using eddy covariance as $F = \overline{\rho}_a\,\overline{w'x'}$, where w is the vertical component of wind velocity, x is CO_2 or DMS mixing ratio, $\overline{\rho}_a$ is dry air density, primes denote fluctuations about the mean value, and the overbar denotes the covariance over the averaging interval [*Kaimal and Finnigan*, 1994].

[6] Two ultrasonic anemometers (CSAT3, Campbell Scientific) separated horizontally by 1 m were mounted on the Knorr's meteorological mast, 2 m forward of the bow and 14 m above the mean sea surface. Gas inlets and a motion sensor (MotionPak2, Systron Donner) were centered between and slightly aft of the sonic anemometer measurement volumes. CO_2 concentrations in air and seawater were measured using 5 InfraRed Gas Analyzers (IRGAs, LiCor, Inc.). The methods used to measure the CO_2 fluxes and piston velocities are described briefly below, and in further detail by S. Miller et al. (Ship-based measurement of air-sea CO_2 exchange by eddy covariance, submitted to *Journal of Geophysical Research*, 2009).

[1] Atmospheric Sciences Research Center, University at Albany, State University of New York, Albany, New York, USA.
[2] Earth System Science, University of California, Irvine, California, USA.
[3] Department of Chemistry, Chapman University, Orange, California, USA.

0094-8276/09/2009GL038907

DMS concentrations and air-sea fluxes were measured using two atmospheric pressure chemical ionization mass spectrometers described by *Marandino et al.* [2007].

[7] The mean atmospheric and seawater CO_2 concentrations were measured continuously in the ship laboratory. Air was drawn from an inlet on the bow mast at 1 slpm through 80 m of 3.9 mm ID tubing to a closed-path IRGA (LI7000). Two additional IRGAs measured the CO_2 concentration of closed-loop air streams equilibrated with the ship's uncontaminated seawater supply: one (LI7000) using a membrane contactor (LiquiCell X40, Knorr-07a and b) and the other (LI820) using a showerhead-type equilibrator (Knorr-07b only). The two systems showed excellent agreement.

[8] Two IRGAs located in the bosun locker directly below the bow mast were used to measure turbulent CO_2 fluctuations (CO_2 IRGA) and H_2O fluctuations (H_2O IRGA). Air was drawn to these IRGAs at 16–18 slpm through an inlet filter near the sonic anemometer and 14 m of 5.9 mm ID tubing, with a travel time of approximately 1 second. Water vapor fluctuations and the associated Webb correction were significantly reduced by drying the airstream using a Nafion drier (PD-200T, PermaPure). This reduced the apparent latent heat flux measured by the downstream CO_2 IRGA by >90%. Laboratory tests indicated that temperature fluctuations (and the associated Webb correction) were negligible due to heat exchange in the sample tube. The CO_2 IRGA was an open-path style sensor (LI7500) converted to a closed-path sensor by inserting a cylindrical cell into the optical path (12.5 cm long, 1.6 cm I.D., volume 0.025 L, flush time less than 0.1 s). This instrument has considerably less motion sensitivity than closed-path sensors (LI7000 and LI6262) previously used for eddy covariance at sea [*McGillis et al.*, 2001]. Fast-response water vapor fluctuations used to calculate latent heat flux were measured by the H_2O IRGA (LI7000), installed upstream of the Nafion drier.

[9] Turbulence data from the sonic anemometer, motion sensor, and CO_2 and H_2O IRGAs in the bosun locker were sampled at 10 Hz, and measured winds were corrected for platform motion [*Miller et al.*, 2008]. Fluxes were calculated using a 13.7-minute averaging period (8192 10 Hz samples). Similar to previous air-sea gas flux studies, flow distortion was addressed by rejecting flux intervals when the mean relative wind direction over the flux interval was >90° from forward (>60° for the quadrant with the adjacent sonic). The turbulent sensible heat and CO_2 fluxes were considered to be stationary when the difference between the 13.7-minute fluxes and the average of the 4 2048 sample (3.4 min) subintervals was less than 30% [*Foken and Wichura*, 1996]. The loss of high-frequency contributions to CO_2 and H_2O flux due to tubing attenuation was corrected using the ogive analysis described by *Marandino et al.* [2007]. The average CO_2 (H_2O) flux loss was 10% (40%) for unstable and 25% (80%) for stable atmospheric conditions. Here, we present k_{CO2} data collected only during unstable atmospheric conditions. The measured turbulent fluxes and air-water concentration differences were used to calculate piston velocities for CO_2 on the Knorr-07a cruise, and for CO_2 and DMS on the Knorr-07b cruise. The three data sets were combined after normalizing the piston velocities to Schmidt number of 660, assuming an $Sc^{-0.5}$ dependence.

3. Results and Discussion

3.1. Overview of Cruises

[10] The cruise tracks and MODIS chlorophyll map for the study region are provided in the auxiliary material.[1] A broad range of chlorophyll concentrations was measured (0.2–3.8, mean 0.8 mg m^{-3}, Figure 1b). Chlorophyll generally increased with latitude. Sea surface temperatures (SST) decreased with latitude and were lowest during the more eastward Knorr-07a cruise (Figure 1a). SST was much warmer than the air temperature south of 52°N during Knorr-07a with ocean to atmosphere (positive) heat flux, slightly warmer than air temperature between ~50–65°N with small net upward heat fluxes, and colder than air temperature south of 50°N during Knorr-07b with net downward heat fluxes. Wind speeds were low to moderate throughout both cruises, with wind at 10-m height range of 1–12 ms^{-1} (Figure 1a).

[11] Data were collected for 326 hours (1304 flux intervals) during Knorr-07a and 200 hours (800 flux intervals) during Knorr-07b. Intervals were rejected based on the following criteria: air-water pCO_2 difference less than 35 ppm (73% of Knorr-07a intervals, 25% of Knorr-07b), instrument spikes (27% Knorr-07a, 32% Knorr-07b), poor wind direction (35% Knorr-07a, 2% Knorr-07b), instationarity of the turbulent CO_2 flux (74% Knorr-07a, 75% Knorr-07b), and stable atmospheric conditions (25% Knorr-07a, 65% Knorr-07b). The resulting high-quality dataset for k_{CO2} included 88 intervals for Knorr-07a intervals and 126 intervals for Knorr-07b. DMS fluxes are reported here during 128 hours during Knorr-07b. Simultaneous high-quality k_{CO2} (40 flux intervals) and k_{DMS} (34 flux intervals) data were obtained between 50°N–54°N latitude (Figure 1d). DMS concentrations and fluxes in the high-chlorophyll region north of 54°N were anomalously high and are not included in our analysis [see *Marandino et al.*, 2008b].

3.2. Air-Sea Concentrations and Fluxes

[12] Atmospheric pCO_2 was relatively constant during each cruise, 387 ± 4 ppm for Knorr-07a and 377 ± 4 ppm for Knorr-07b. Sea surface pCO_2 was undersaturated with respect to the atmosphere, except for the last part of Knorr-07b, when it was supersaturated. The mean water-air pCO_2 difference was −49 ± 22 ppm during undersaturated periods north of 39°N, and 43 ± 28 ppm in the supersaturated region (Figure 1b). Surface waters were supersaturated with DMS throughout Knorr-07b, with a mean concentration of 2.5 ± 1.1 nM (Figure 1b) [*Marandino et al.*, 2008b].

[13] The direction of the turbulent fluxes of CO_2 and DMS was consistent with their respective water-air partial pressure gradients (Figures 1b and 1c). The average downward CO_2 flux was −3.3 ± 0.1 mol CO_2 m^{-2} yr^{-1} in the ocean sink region, and the average upward flux was 1.7 ± 1 mol CO_2 m^{-2} yr^{-1} in the source area. DMS fluxes were from ocean to atmosphere, and averaged 5.7 ± 0.4 μmol

[1] Auxiliary material data sets are available at ftp://ftp.agu.org/apend/gl/2009gl038907. Other auxiliary material files are in the HTML.

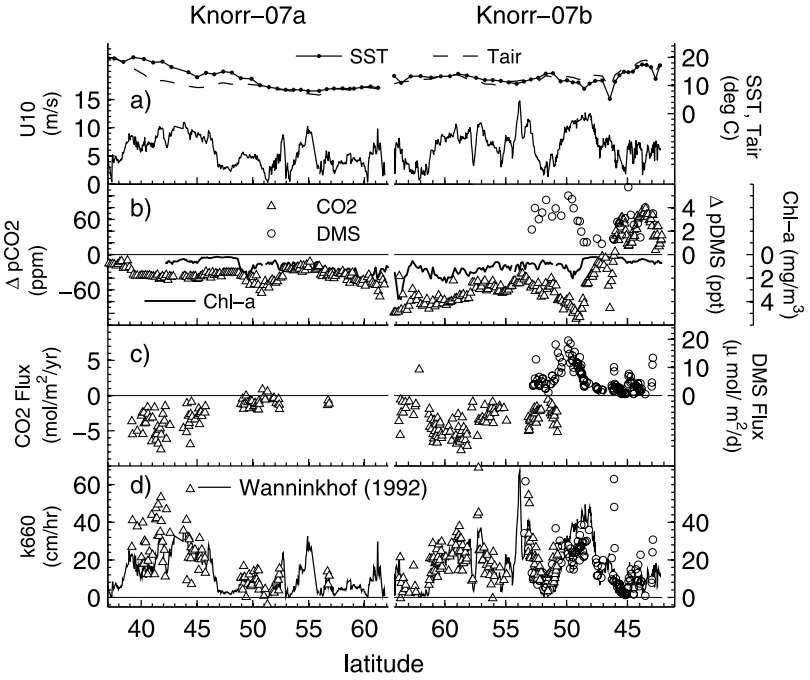

Figure 1. Summary of Knorr-07 cruises. The left side shows latitude increasing, Bridgetown, Barbados to Reykjavik, Iceland (29 May-9 Jun); the right side shows latitude decreasing, Reykjavik, Iceland to Woods Hole, MA (17–26 Jul). (a) Wind speed at 10 m height (m s^{-1}, solid), SST (solid with dots, deg C) and air temperature (dashed, deg C). (b) Air-water concentration difference for pCO_2 (ppm, triangles) and pDMS (ppt, circles), and MODIS chlorophyll (mg m^{-3}, solid curve) along the cruise track from the 17 May–17 Jun, 2007 image (on the left) and the Jul 2007 monthly image (on the right). (c) Air-sea flux of CO_2 (mol CO_2 m^{-2} yr^{-1}, triangles) and DMS (μmol m^{-2} d^{-1}, circles), positive values are from ocean to atmosphere; and d) k_{CO2} (triangles) and k_{DMS} (circles) in cm hr^{-1} for Sc = 660, along with parameterization of *Wanninkhof* [1992] (solid curve).

m^{-2} d^{-1} for measurements south of 54°N included in the analysis.

3.3. Gas Transfer Coefficients

[14] The piston velocities for CO_2 and DMS measured on the Knorr-07 cruises were remarkably similar (Figure 2). k_{CO2} and k_{DMS} increased with wind speed and exhibited scatter typical of air-sea flux measurements. The measured piston velocities were mostly higher than the quadratic relationship of *Wanninkhof* [1992]. The linear regression of the combined CO_2 and DMS data (n = 323) against 10-m wind speed was $k_{660} = 3.0U_{10} - 2.5$ cm · hr^{-1}, $r^2 = 0.57$. Nineteen data points (6% of the data set) were flagged as outliers using the 95% confidence interval of the residuals for the linear fit. The regression slope differed by less than 10% and the r^2-statistic decreased to 0.45 when the outlier points were included. The r^2-statistic did not improve when a quadratic instead of linear fit was assumed. The r^2-statistic was also similar for linear and quadratic regressions calculated independently for CO_2 and DMS. To our knowledge, these are the first open-ocean eddy covariance CO_2 data suggesting a linear relation between wind speed and piston velocity at low winds. For DMS, the four EC data sets published to date are consistent with a linear k versus U_{10} relationship [*Marandino et al.*, 2008a]. This study, and the previous DMS studies cover a broad range of oceanographic regions and environmental conditions.

[15] Our results are markedly different than those from the GasEx-1998 and GasEx-2001 experiments (Figure 3a).

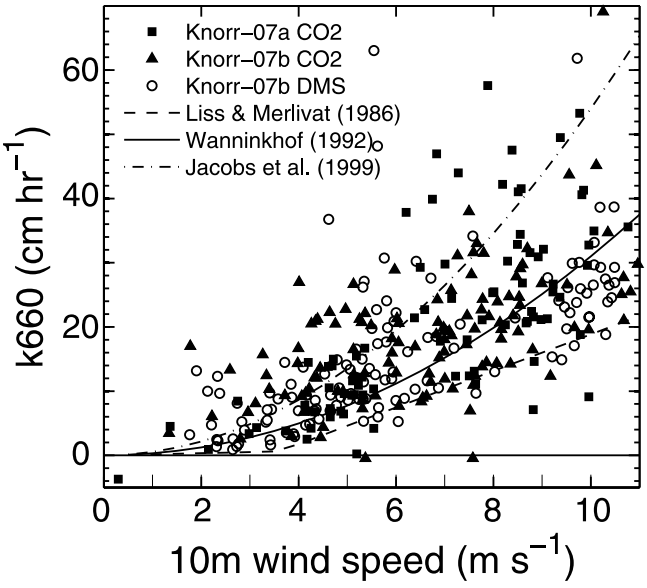

Figure 2. Piston velocity (cm hr^{-1}) for Sc = 660 versus 10-m wind speed: Knorr-07a k_{CO2} (squares), Knorr-07b k_{CO2} (triangles) and k_{DMS} (circles), *Liss and Merlivat* [1986] (dashed), *Wanninkhof* [1992] (solid), and *Jacobs et al.* [1999] (dash-dotted).

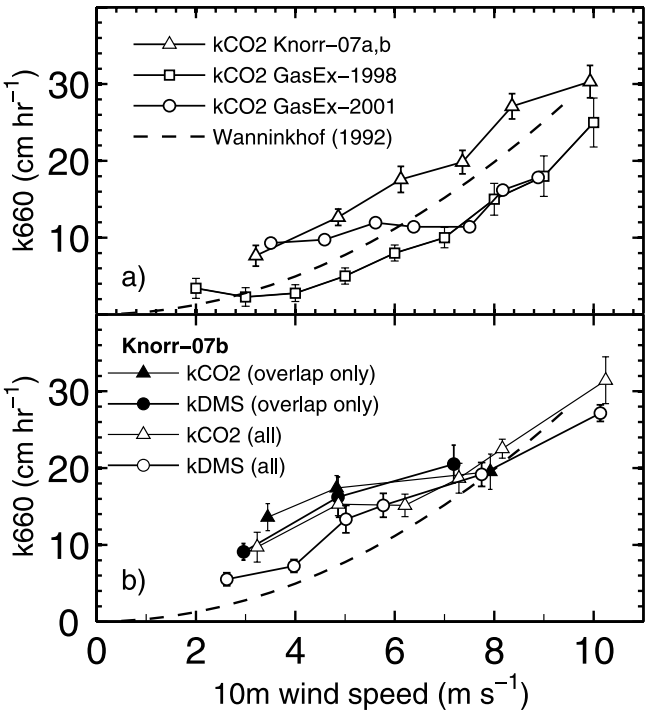

Figure 3. Piston velocity (cm hr^{-1}) versus 10-m wind speed for Sc = 660. Outliers were removed using a 99% confidence interval before calculating bin averages. (a) Eddy covariance CO_2-based measurements: North Atlantic Knorr-07a,b cruises (triangles), North Atlantic GasEx-1998 (squares), Eastern Tropical Pacific GasEx-2001 (circles), and parameterization of *Wanninkhof* [1992] (dashed). Vertical bars represent standard error. For Gas Ex-1998, standard error was estimated using published standard deviations [*McGillis et al.*, 2001], and estimated number of samples in each wind speed bin based on the total experiment duration, flux averaging period, and assuming a Rayleigh wind speed distribution. (b) Knorr-07b only CO_2 (triangles) and DMS (circles). Open symbols correspond to the entire Knorr-07b cruise dataset, and filled symbols correspond to the latitude range 50°N–54°N with simultaneous CO_2 and DMS data (see Figure 1d).

GasEx-1998 in the North Atlantic found very low piston velocities at low wind speed, leading *Wanninkhof and McGillis* [1999] to hypothesize that biologically produced surfactants suppressed gas transfer. In contrast, the Equatorial Pacific GasEx-2001 experiment found very high piston velocities at low wind speed, attributed to turbulence caused by diurnally varying heating of the surface ocean. The Knorr-07 cruises traversed waters of both high and low chlorophyll levels (Figure 1b), and presumably encountered waters with a wide range of biological activity and biologically-generated surfactants. The Knorr-07 k_{CO2} values are significantly larger than the two GasEx data sets and are slightly larger than the *Wanninkhof* [1992] parameterization (Figure 3a). It is not possible to identify a single cause for the differences between our results and those from the GasEx studies. Multiple factors such as sea state, heat flux, and microlayer thickness may all play a role. Methodological differences between the two studies are also possible.

[16] The DMS solubility was roughly 20 times that of CO_2 during the Knorr-07 cruises. Theory suggests that breaking waves and bubbles can enhance the transfer of lower solubility gases relative to higher solubility gases [*Woolf*, 1997]. Gas transfer models that include wind speed and wave state have been developed [*Soloviev et al.*, 2007]. However, measurements of sea state have generally not been reported along with direct gas flux measurements, and wave state effects have been inferred from wind speed measurements alone. For the range of wind speeds measured during the Knorr-07 cruises, the whitecap coverage was expected to be small (0.01–1%) [*Monahan and Muircheartaigh*, 1980]. The data-based model of *Asher and Wanninkhof* [1998] predicts only a few percent enhancement of k_{CO2} relative to k_{DMS} for $U_{10} < 10$ ms^{-1}. The data from Knorr-07b show that the measured k_{CO2} and k_{DMS} were similar over the entire wind speed range (Figure 3b). The similarity between k_{CO2} and k_{DMS} was found both for the entire Knorr-07b dataset and for the latitude range 50°N–54°N where simultaneous CO_2 and DMS data were obtained (Figures 1d and 3b, solid symbols). The data confirm that solubility did not play a major role in gas transfer over the wide range of oceanographic conditions encountered on this cruise.

4. Conclusion

[17] Understanding gas transfer at low-moderate wind speeds is important since winds less than 10 ms^{-1} cover more than 60% of the oceans on average. Eddy covariance piston velocities measured for CO_2 and DMS during low-moderate wind speed on two North Atlantic cruises were remarkably similar, exhibiting a linear relationship to wind speed. Wind speed accounted for over half of the variance of the gas transfer coefficient. The wind speed-dependence of these data is quite different than those from earlier CO_2-based studies, but similar to prior DMS-based studies. The similarity between the k versus U_{10} relationships for CO_2 and DMS suggests that solubility was not a significant factor under these low to moderate wind conditions. The Knorr-07 data set is relatively limited in terms of the overall number of flux measurements, the temporal overlap between the CO_2 and DMS flux measurements, and the wind speed range. However, it demonstrates the utility of this approach as a means of improving our understanding of air-sea gas transfer.

[18] **Acknowledgments.** Thanks to Liz Caporelli, Captains Kent Sheasley and George Silva, the WHOI support staff and R/V Knorr crew, Cyril McCormick, Brandon Finley, Mike Lawler, Jim Albrecht, and the Ministry for Foreign Affairs of Iceland. This work was supported by NSF Atmospheric Chemistry (ATM0426314) and is a contribution to US SOLAS. MODIS data from OceanColor web site (http://oceancolor.gsfc.nasa.gov).

References

Asher, W. E., and R. Wanninkhof (1998), The effect of bubble-mediated gas transfer on purposeful dual-gaseous tracer experiments, *J. Geophys. Res.*, *103*(C5), 10,555–10,560, doi:10.1029/98JC00245.

Blomquist, B. W., C. W. Fairall, B. J. Huebert, D. J. Kieber, and G. R. Westby (2006), DMS sea-air transfer velocity: Direct measurements by eddy covariance and parameterization based on the NOAA/COARE gas transfer model, *Geophys. Res. Lett.*, *33*, L07601, doi:10.1029/2006GL025735.

Broecker, W. S., and W. Siems (1984), The role of bubbles for gas transfer from water to air at higher windspeeds. Experiments in the

wind-wave facility in Hamburg, in *Gas Transfer at Water Surfaces*, edited by W. Brutsaert and G. H. Jirka, pp. 229–238, D. Reidel, Dordrecht, Netherlands.

Foken, T., and B. Wichura (1996), Tools for quality assessment of surface-based flux measurements, *Agric. For. Meteorol.*, *78*(1–2), 83–105, doi:10.1016/0168-1923(95)02248-1.

Hare, J. E., C. W. Fairall, W. R. McGillis, J. B. Edson, B. Ward, and R. Wanninkhof (2004), Evaluation of the National Oceanic and Atmospheric Administration/Coupled-Ocean Atmospheric Response Experiment (NOAA/COARE) air-sea gas transfer parameterization using GasEx data, *J. Geophys. Res.*, *109*, C08S11, doi:10.1029/2003JC001831.

Ho, D. T., C. S. Law, M. J. Smith, P. Schlosser, M. Harvey, and P. Hill (2006), Measurements of air-sea gas exchange at high wind speeds in the Southern Ocean: Implications for global parameterizations, *Geophys. Res. Lett.*, *33*, L16611, doi:10.1029/2006GL026817.

Huebert, B. J., B. W. Blomquist, J. E. Hare, C. W. Fairall, J. E. Johnson, and T. S. Bates (2004), Measurement of the sea-air DMS flux and transfer velocity using eddy correlation, *Geophys. Res. Lett.*, *31*, L23113, doi:10.1029/2004GL021567.

Jacobs, C., W. Kohsiek, and W. Oost (1999), Air-sea fluxes and transfer velocity of CO_2 over the North Sea: results from ASGAMAGE, *Tellus, Ser. B*, *51*(3), 629–641, doi:10.1034/j.1600-0889.1999.t01-2-00005.x.

Jahne, B., and H. Haussecker (1998), Air-water gas exchange, *Annu. Rev. Fluid Mech.*, *30*(1), 443–468, doi:10.1146/annurev.fluid.30.1.443.

Kaimal, J. C., and J. J. Finnigan (1994), *Atmospheric Boundary Layer Flows*, Oxford Univ. Press, New York.

Liss, P. S., and L. Merlivat (1986), Air-sea gas exchange rates: Introduction and synthesis, in *The Role of Air-Sea Exchange in Geochemical Cycling*, *NATO ASI Ser., Ser. C*, vol. 185, edited by P. Buart-Ménard, pp. 113–127, D. Reidel, Dordrecht, Netherlands.

Marandino, C. A., W. J. De Bruyn, S. D. Miller, and E. S. Saltzman (2007), Eddy correlation measurements of the air/sea flux of dimethylsulfide over the North Pacific Ocean, *J. Geophys. Res.*, *112*, D03301, doi:10.1029/2006JD007293.

Marandino, C. A., W. J. De Bruyn, S. D. Miller, and E. S. Saltzman (2008a), Open ocean DMS air/sea fluxes over the eastern South Pacific Ocean, *Atmos. Chem. Phys. Discuss.*, *8*, 12,081–12,114.

Marandino, C. A., W. J. De Bruyn, S. D. Miller, and E. S. Saltzman (2008b), DMS air/sea flux and gas transfer coefficients from the North Atlantic summertime coccolithophore bloom, *Geophys. Res. Lett.*, *35*, L23812, doi:10.1029/2008GL036370.

McGillis, W. R., J. B. Edson, J. E. Hare, and C. W. Fairall (2001), Direct covariance air-sea CO_2 fluxes, *J. Geophys. Res.*, *106*(C8), 16,729–16,746, doi:10.1029/2000JC000506.

McGillis, W. R., et al. (2004), Air-sea CO_2 exchange in the equatorial Pacific, *J. Geophys. Res.*, *109*, C08S02, doi:10.1029/2003JC002256.

Miller, S. D., T. S. Hristov, J. B. Edson, and C. A. Friehe (2008), Platform motion effects on measurements of turbulence and air-sea exchange over the open ocean, *J. Atmos. Oceanic Technol.*, *25*, 1683–1694, doi:10.1175/2008JTECHO547.1.

Monahan, E. C., and I. Muircheartaigh (1980), Optimal power-law description of oceanic whitecap coverage dependence on wind speed, *J. Phys. Oceanogr.*, *10*(12), 2094–2099, doi:10.1175/1520-0485(1980)010<2094:OPLDOO>2.0.CO;2.

Nightingale, P. D., G. Malin, C. S. Law, A. J. Watson, P. S. Liss, M. I. Liddicoat, J. Boutin, and R. C. Upstill-Goddard (2000), In situ evaluation of air-sea gas exchange parameterizations using novel conservative and volatile tracers, *Global Biogeochem. Cycles*, *14*(1), 373–387, doi:10.1029/1999GB900091.

Soloviev, A., M. Donelan, H. Graber, B. Haus, and P. Schlüssel (2007), An approach to estimation of near-surface turbulence and CO_2 transfer velocity from remote sensing data, *J. Mar. Syst.*, *66*(1–4), 182–194, doi:10.1016/j.jmarsys.2006.03.023.

Wanninkhof, R. (1992), Relationship between wind speed and gas exchange over the ocean, *J. Geophys. Res.*, *97*(C5), 7373–7382, doi:10.1029/92JC00188.

Wanninkhof, R., and W. R. McGillis (1999), A cubic relationship between air-sea CO_2 exchange and wind speed, *Geophys. Res. Lett.*, *26*(13), 1889–1892, doi:10.1029/1999GL900363.

Wanninkhof, R., J. Ledwell, and W. S. Broecker (1985), Gas exchange-wind speed relation measured with sulfur hexafluoride on a lake, *Science*, *227*(4691), 1224–1226, doi:10.1126/science.227.4691.1224.

Woolf, D. K. (1997), Bubbles and their role in gas exchange, *The Sea Surface and Global Change*, edited by P. S. Liss and R. A. Duce, pp. 173–205, Cambridge Univ. Press, Cambridge, U. K.

W. de Bruyn, Department of Chemistry, Chapman University, Orange, CA 92866, USA.

C. Marandino and E. S. Saltzman, Earth System Science, University of California, Irvine, CA 92697-3100, USA.

S. Miller, Atmospheric Sciences Research Center, University at Albany, State University of New York, 251 Fuller Road L317, Albany, NY 12203, USA.